

# *Determination of Unfiltered In-Leakage by ATD and AIMS E 741 Techniques*

# Russell N. Dietz, Head

Tracer Technology Center  
Brookhaven National Laboratory  
Upton, New York 11973

Dietz@bnl.gov

(631) 344-3059

<http://www.ecd.bnl.gov/TTC.html>

Brookhaven Science Associates  
U.S. Department of Energy



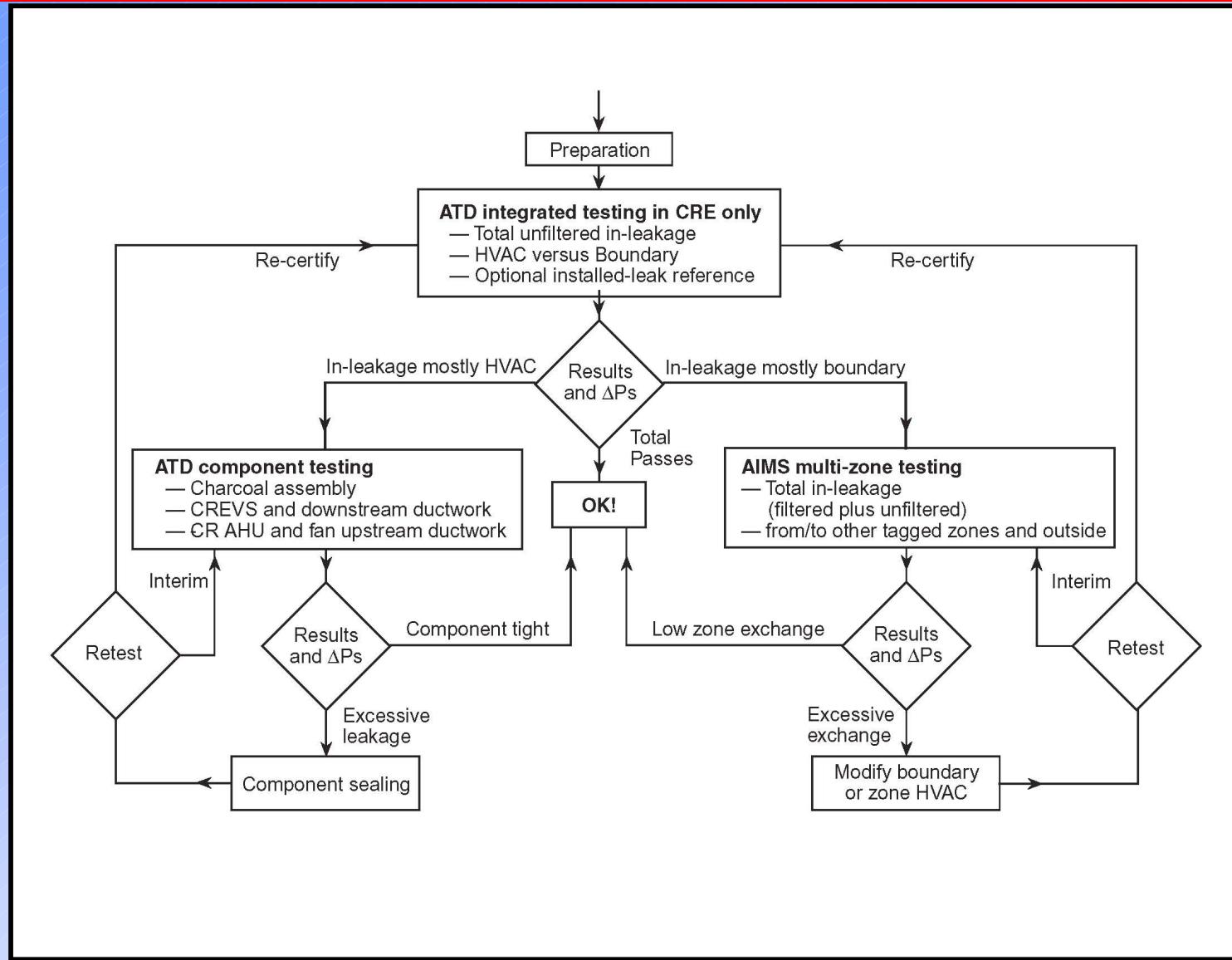
# Control Room In-Leakage by ATD and AIMS Techniques

---

## Outline

-  Suggested industry approach
-  Brief look at the 4 tracer techniques
  - Applicability of E 741 to all 4
- 3. AIMS (Air Infiltration Measurement System)
  - Calvert Cliff results and uncertainties
- 4. New ATD (Atmospheric Tracer Depletion)
  - Mathematics of the direct measurement of unfiltered in-leakage
  - Results of laboratory tests
  - Expected uncertainties
- 5. Current status
  - Audit by Dominion Energy of QAP (8/4 –8/03)
  - Testing at 3 Dominion stations and Pilgrim Nuclear
- 6. Special issues regarding contracting to a National Laboratory

# Suggested Industry Approach Using ATD and AIMS



# Applicability of ASTM E 741 to Four Techniques for Measuring Unfiltered In-Leakage

Major elements in E 741: 18  
- Specifically applicable: 14<sup>a</sup>  
Total specific sub-elements: 108

Technique	SF <sub>6</sub> Decay	SF <sub>6</sub> Inject	AIMS	ATD
<i>Conforms to specific sub-elements:</i>	81	83	84	66
(percentage of subtotal)	(89%)	(89%)	(91%)	(92%)
- does not conform	9	9	8	5
- uncertain (element 3.1.7.1) <sup>b</sup>	1	1	--	1
Subtotal:	91	93	92	72 <sup>c</sup>
<i>Not applicable:</i>	17	15	16	36
<b>Direct unfiltered in-leakage:</b>	No	No	No	Yes
<b>Individual components testable:</b>				
- easily	No	No	No	Yes
- with special tagging	No	Yes	Yes	na
- defined in E 741	No	No	No	No

<sup>a</sup> Others are general to all 4 techniques or to none

<sup>b</sup> Adequately handles single and multi-zone CREs within multi-zone facilities

<sup>c</sup> Lower number of sub elements due to no need for tracer injection.

## APPENDIXES

## (Nonmandatory Information)

**X1. TRACER GASES USED TO DETERMINE AIR CHANGE**

X1.1 Tables X1.1 and X1.2 list tracer gases that are used to determine air change. They include only those gases that have

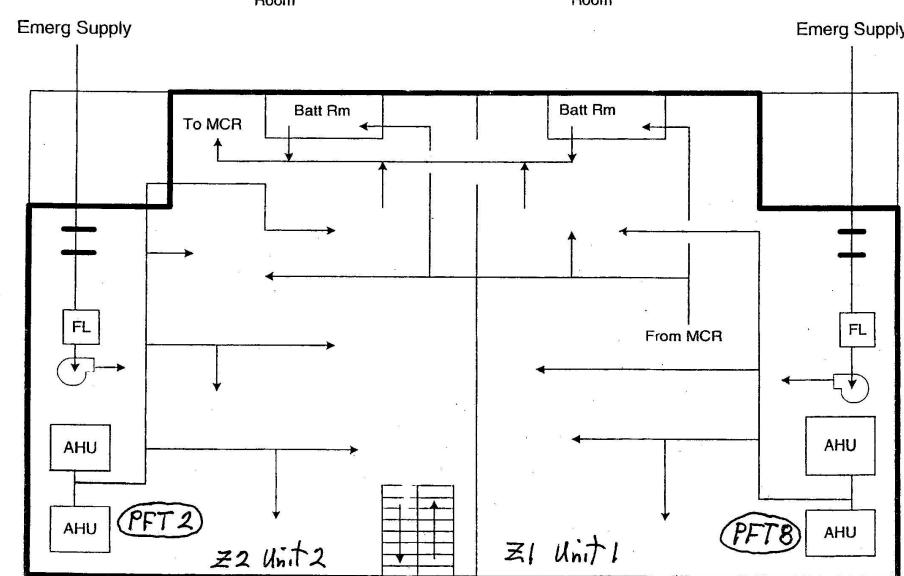
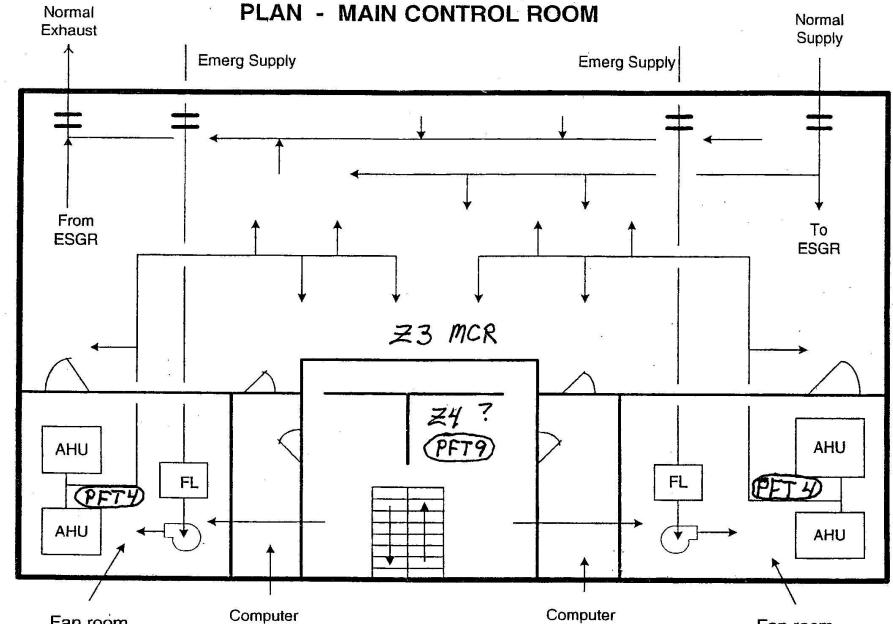
an established personal exposure limit (PEL, see Section 7). Table X1.1 describes the safety properties of gases. Table X1.2 highlights the concentration analysis of gases.

**TABLE X1.1 Tracer Gases and Safety Issues**

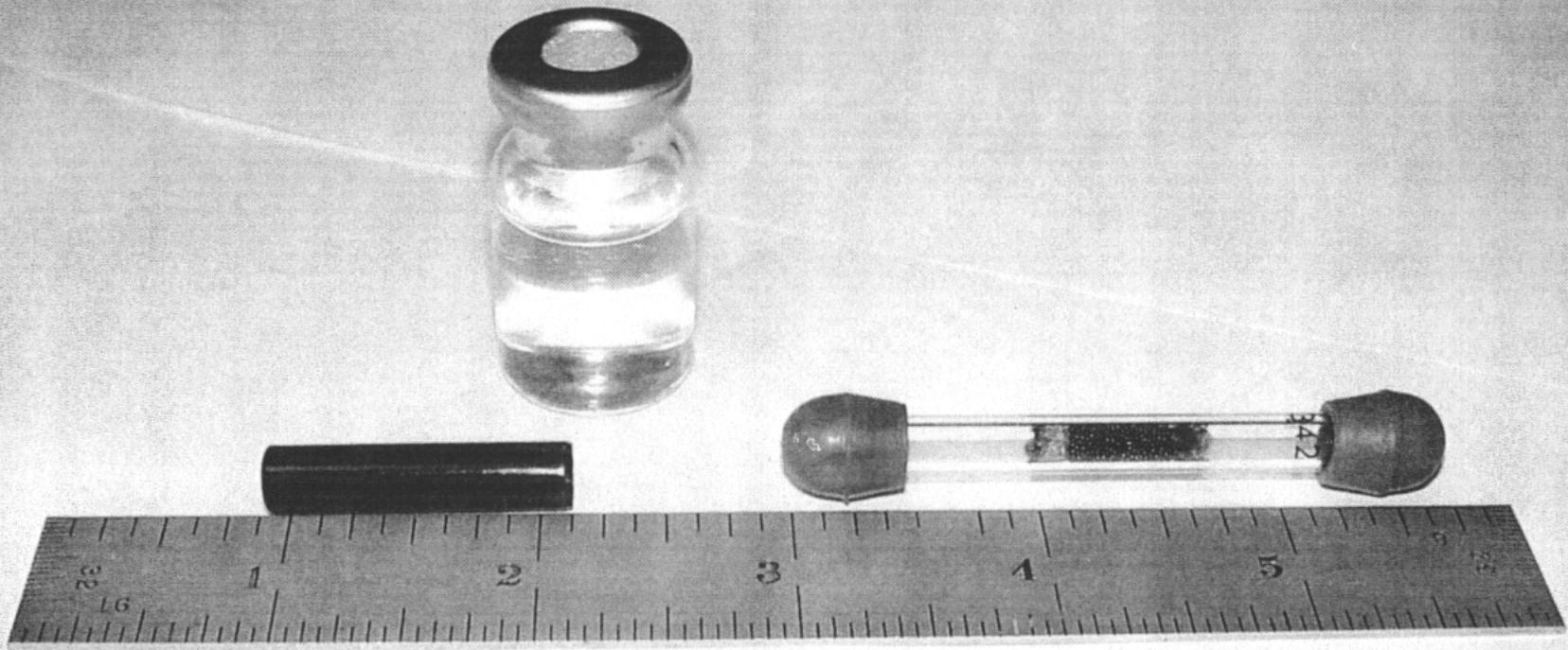
Tracer Gas	PEL	Toxicology	Chemical Reactivity	Comments
Hydrogen	Asphyxiant	Non-toxic	Highly reactive in presence of heat, flame, or O <sub>2</sub>	Fire and explosion hazard when exposed to heat, flame, or O <sub>2</sub>
Helium	Asphyxiant	Non-toxic	Inert	
Carbon Monoxide	50 ppm	Combines with hemoglobin to cause anoxia	Highly reactive with O <sub>2</sub>	Fire and explosion hazard when exposed to heat or flame
Carbon Dioxide	5000 ppm	Can be eye irritant	Reacts vigorously with some metals; soluble in water	
Sulfur Hexafluoride	1000 ppm	Non-toxic	Inert	Thermal decomposition yields highly toxic compounds
Perfluorocarbon tracers (PFTs) <sup>1</sup>	Asphyxiant	Non-toxic	Inert	Thermal decomposition may produce toxic compounds
Nitrous Oxide	25 ppm	Moderately toxic by inhalation	Violent reaction with aluminum; water soluble	Can form explosive mixture with air; ignites at high temperature
Ethane	Asphyxiant	Non-toxic	Flammable	Incompatible with chlorine and oxidizing materials
Methane	Asphyxiant	Non-toxic	Flammable	Incompatible with halogens and oxidizing materials
Octafluorocyclobutane (Halocarbon C-318)	1000 ppm	Low toxicity	Nonflammable	Thermal decomposition yields highly toxic compounds
Bromotrifluoromethane (Halocarbon 13B1)	500 ppm	Moderately toxic by inhalation	Incompatible with aluminum	Dangerous in a fire
Dichlorodifluoromethane (Halocarbon 12)	1000 ppm	Central nervous system and eye irritant; can be narcotic at high levels	Nonflammable; can react violently with aluminum	Thermal decomposition yields highly toxic fumes
Dichlorotetrafluoromethane (Halocarbon 116)	1000 ppm	Can be asphyxiant, mildly irritating, narcotic at high levels	Can react violently with aluminum	Thermal decomposition yields highly toxic fumes

<sup>1</sup> There are a family of 9 PFTs available for multi-zone testing.

NORTH ANNA CRE VENTILATION  
PLAN - MAIN CONTROL ROOM



PLAN - EMERGENCY SWITCHGEAR ROOM



# PFT Testing at Calvert Cliffs

---

## Benchmark Results

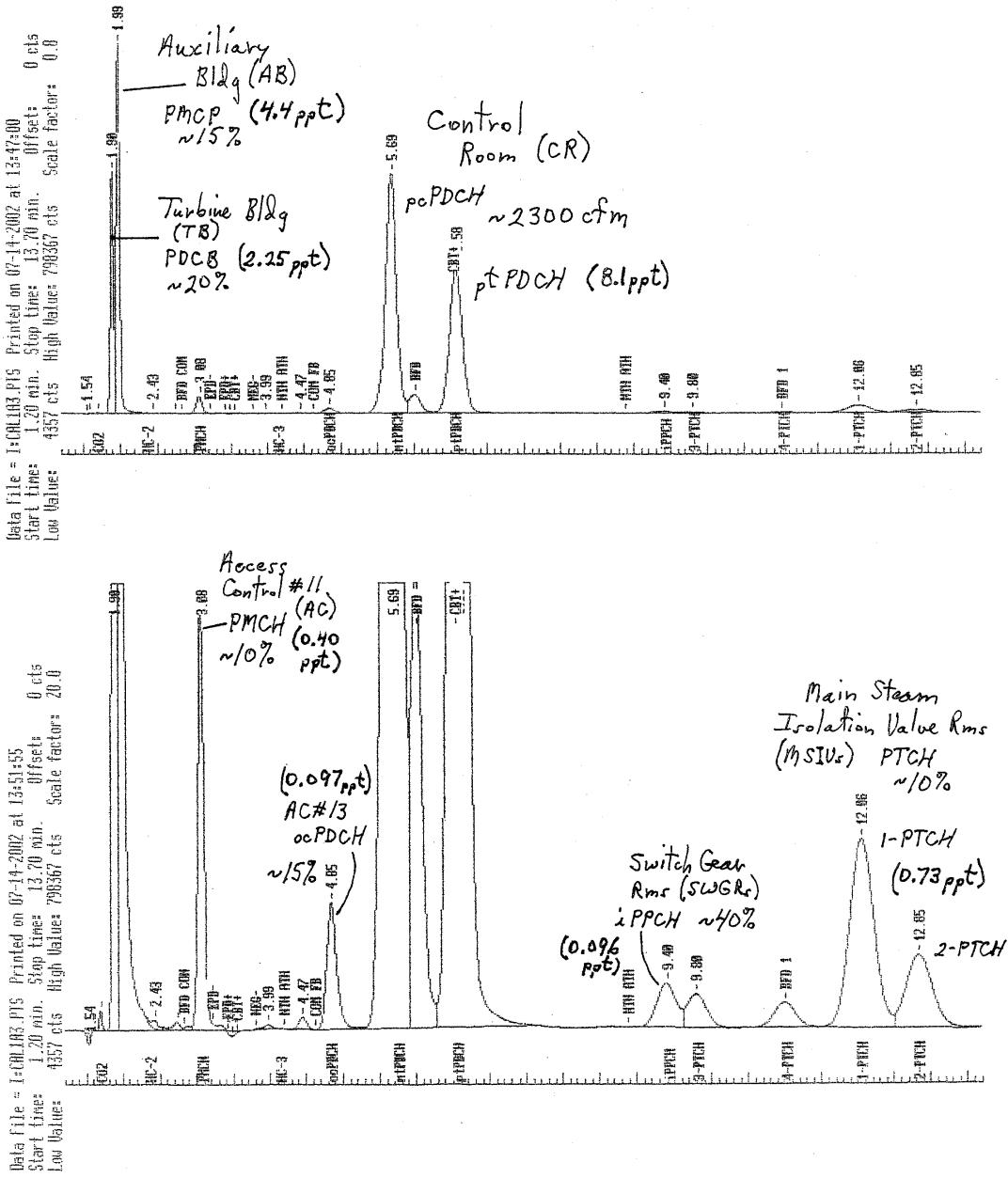
SF6 Test Data (Jan 2000)

- ❖ 11 Train – 3000 CFM +/- 250 CFM
- ❖ 12 Train – 2600 CFM +/- 200 CFM

PFT Test Data (June 2002)

- ❖ 11 Train – 2930 CFM +/- 185 CFM

## MULTI-TRACER CONTROL ROOM AIR INLEAKAGE

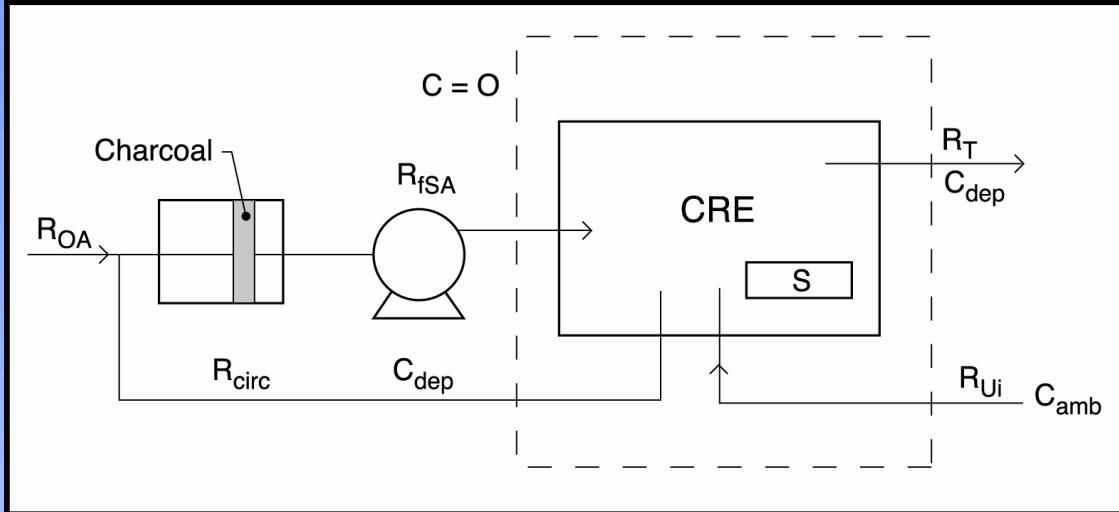


# Calvert Cliffs Total Inleakage was $2930 \pm 185$ cfm. Other flows, in cfm, were:

---

Zone	From/To	CR Inleakage	% of total	CR Outleakage	% of total
0	Outside	$275 \pm 185$	9	$1966 \pm 470$	64
2	AB	$436 \pm 157$	15	$366 \pm 248$	13
3	TB	$466 \pm 172$	16	$599 \pm 415$	20
4	MSIVs	$272 \pm 134$	9	$44 \pm 33$	2
5	AC11	$274 \pm 33$	9	$19 \pm 3$	1
6	AC13	$387 \pm 38$	13	$11 \pm 8$	0
7	SWGRs	$818 \pm 114$	28	$21 \pm 10$	1

# Comparison of E 741 ATD and Injection Mathematics



Material balance around CRE:

E 741 ATD:

$$R_{fSA} + R_{ui} = R_T + R_{circ}$$

$$C = 0 \quad C_{amb} \quad C_{dep} \quad C_{dep}$$

$$S_{in} = S_{out} = R \times C$$

$$\begin{aligned} R_{ui} C_{amb} &= C_{dep} (R_T + R_{circ}) \\ &= C_{dep} (R_{fSA} + R_{Ui}) \end{aligned}$$

$$R_{ui} (C_{amb} - C_{dep}) = R_{fSA} C_{dep}$$

$$\begin{aligned} R_{ui} &= R_{fSA} C_{dep} / (C_{amb} - C_{dep}) \\ &= R_{fSA} / (C_{amb}/C_{dep} - 1) \end{aligned}$$

E 741 injection:

(after charcoal saturated with  $SF_6$ )

$$R_{OA} + R_{ui} = R_T$$

$$C = 0 \quad C.0 \quad C_S$$

$$S_{in} = S_{out} = R \times C$$

$$S = R_T C_S$$

$$R_T = S/C_S$$

$$R_{ui} = S/C_S - R_{OA}$$

## VTMX Chromatogram

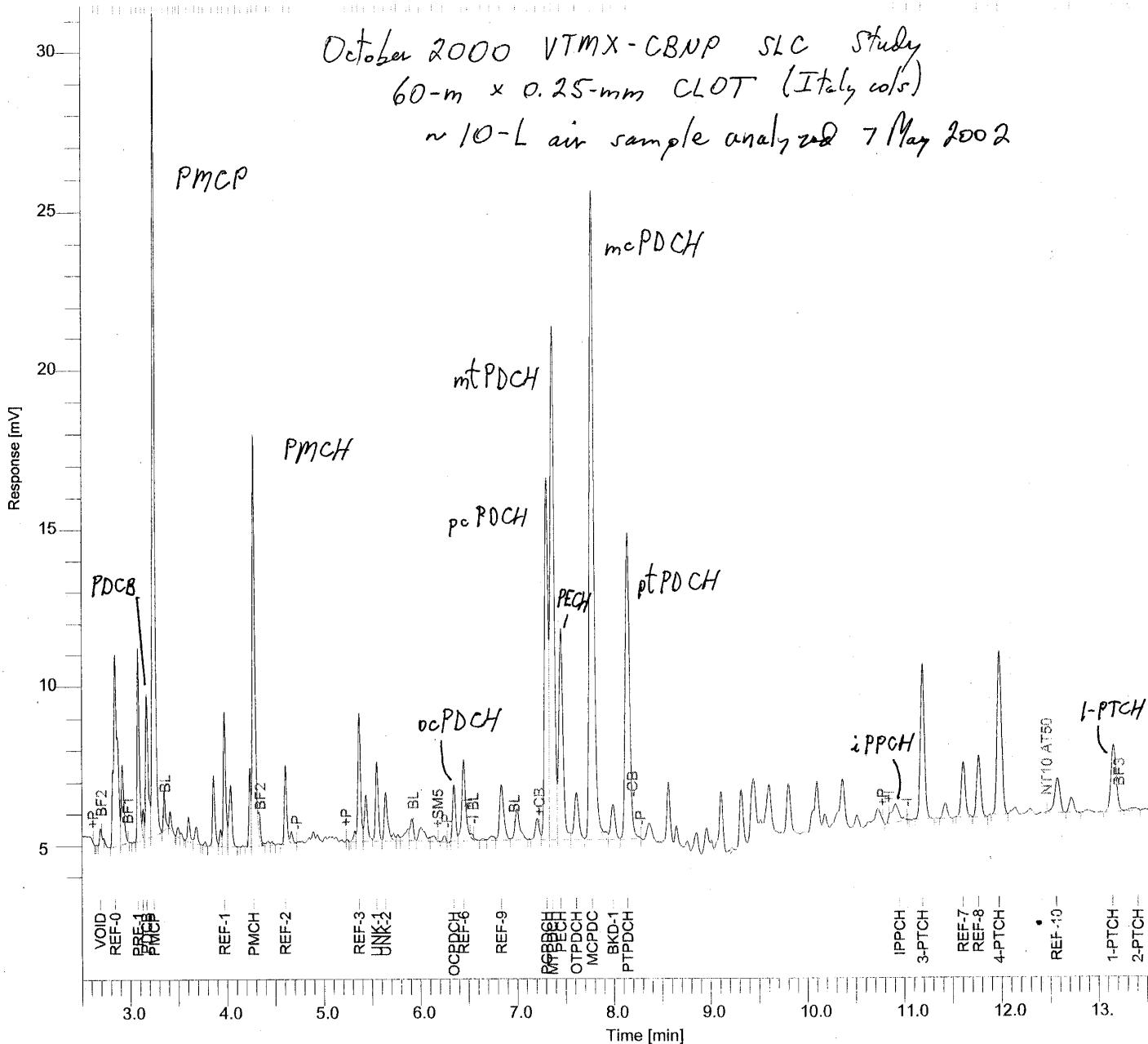
Sample Name : L0007  
File Name : C:\Data\2002\VTMX\Jerome(8)\L0006\_L0007\_L0008\_L0009\_030.raw  
Date : 7/15/02 7:41:25 AM  
Method : 60cps.a(VTMX)copy.mth  
Start Time : 2.50 min End Time : 13.50 min  
Plot Offset: 3.29 mV Plot Scale: 28.0 mV

Sample #: 06

Page 1 of 1

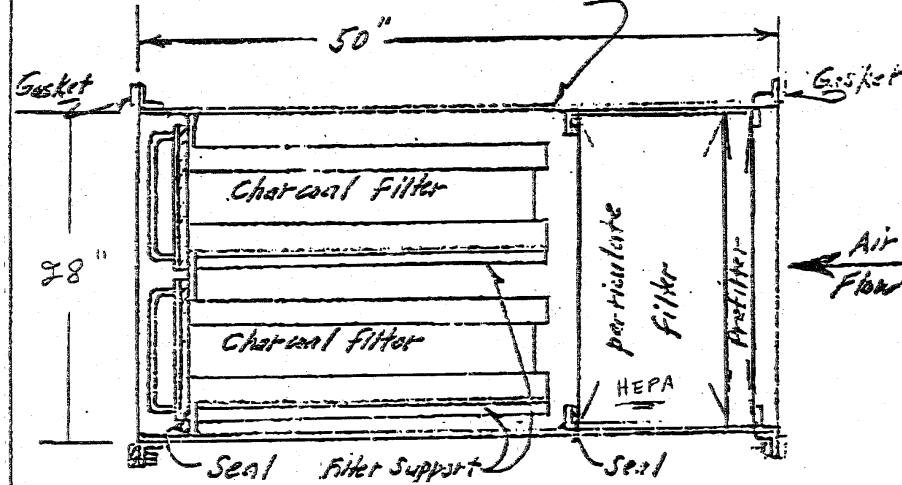
Time of Injection: 5/7/02 8:24:13 PM

Low Point : 3.29 mV High Point : 31.26 mV

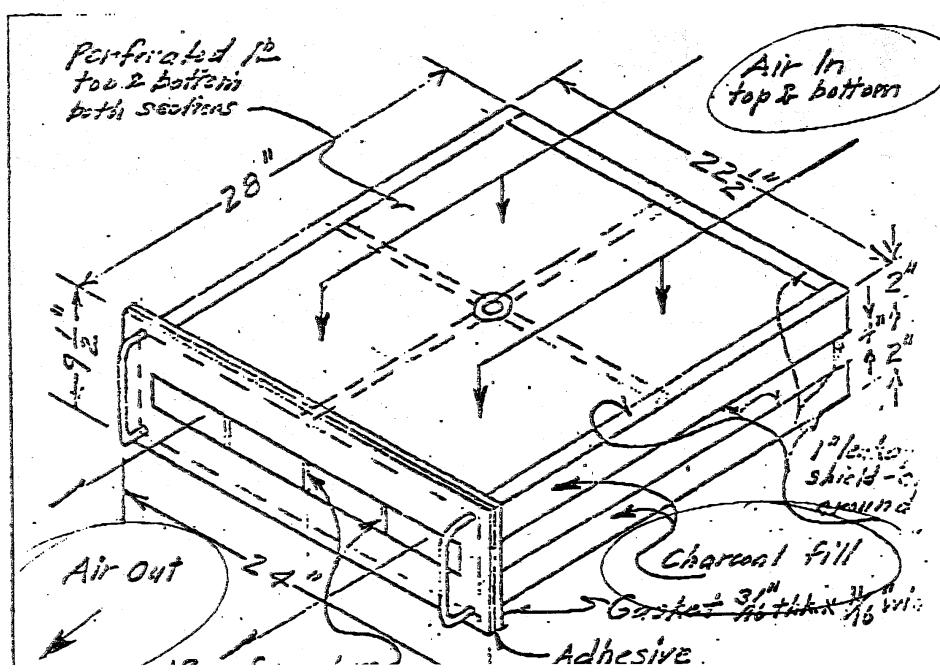


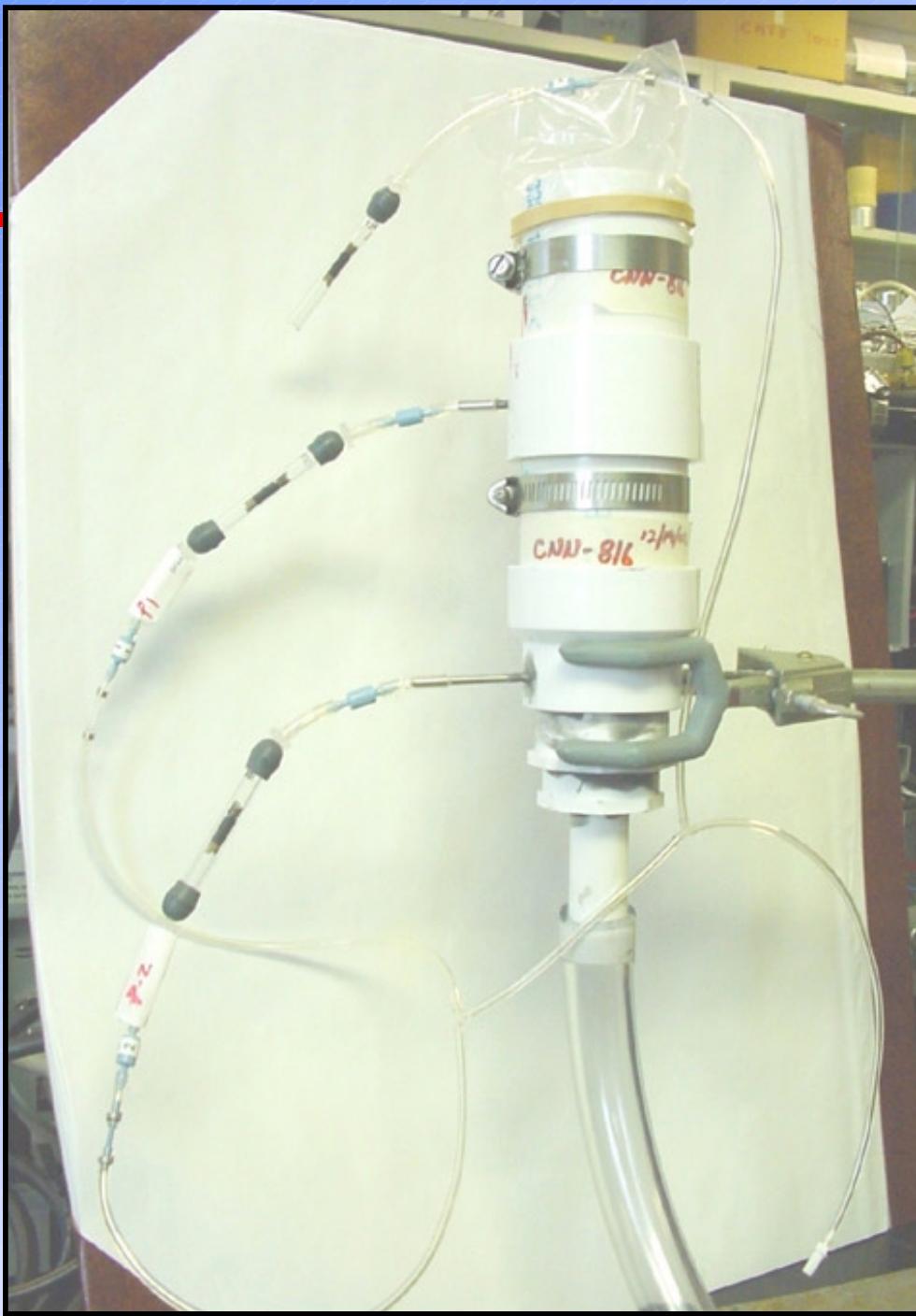
### Filter Assembly - Side View

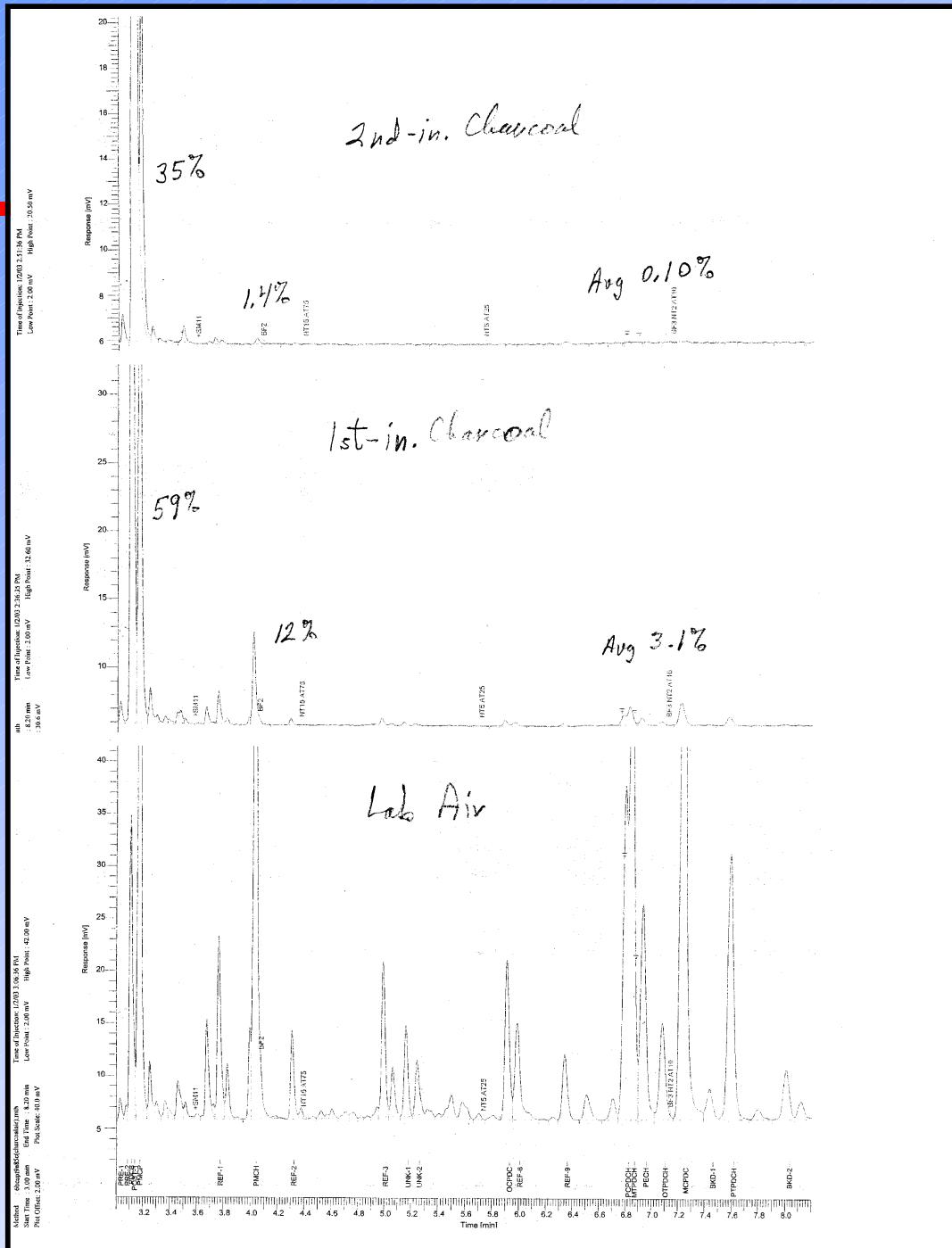
16 gage casing



### CHARCOAL CELL







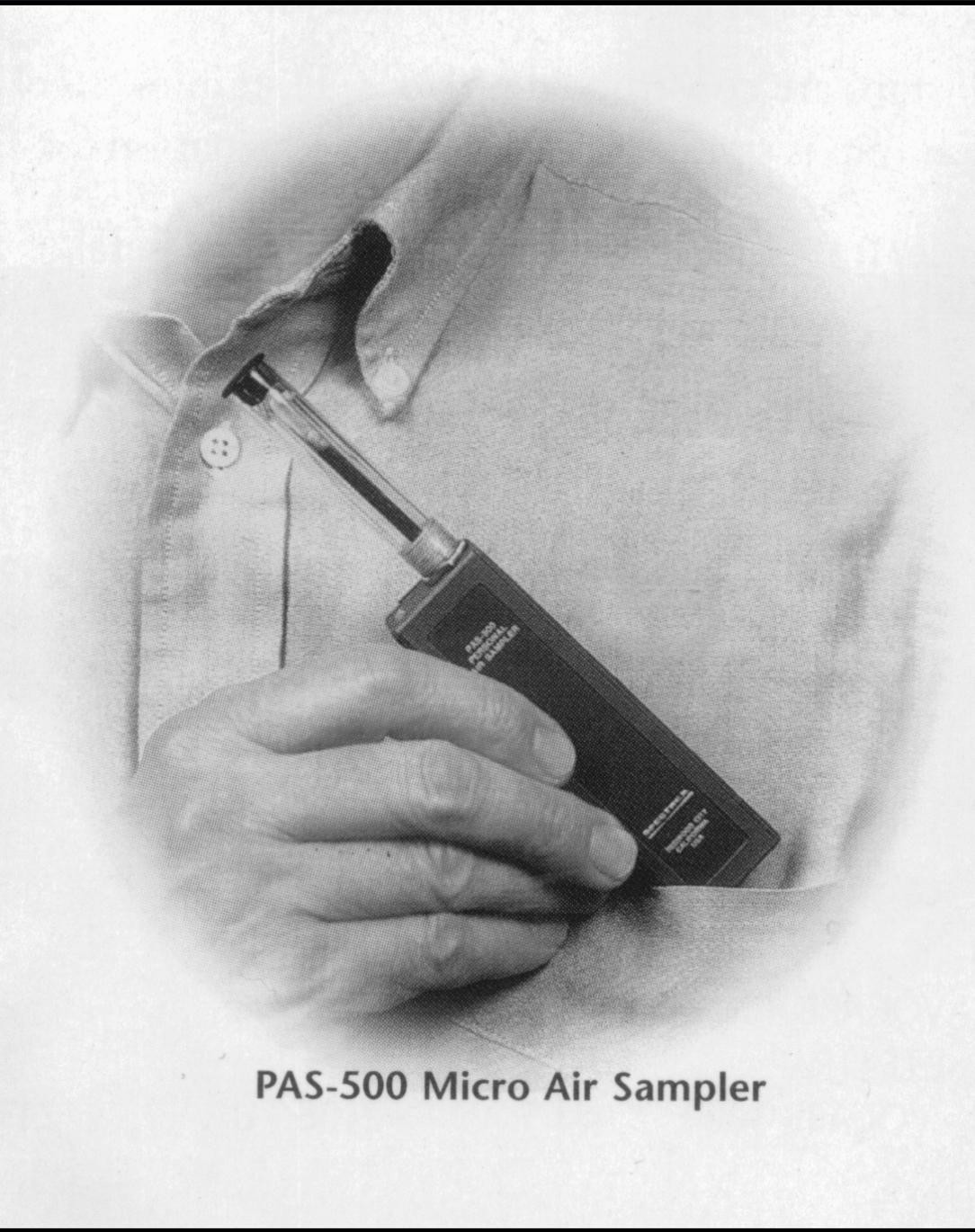
# Example Precision of Atmospheric PFT Background Measurements

---

(taken from October 2000 Salt Lake City – SLC -- tests)

- ❖ ATD control room envelope sample volumes will be ~18 to 50 L
  - ❖ -- depleted concentrations to be measured down to 1 to 3% remaining
- ❖ SLC sample volumes were ~0.5 L (~3% of CRE sample vol.)
  - ❖ -- area ratios used to normalize for variable sample volumes between 480 sampling pumps

Expected ATD Precision (SLC PFT Peak Area Ratios)			
	pc/pt	mt/pt	mc/pt
Area ratio:	0.918	1.524	2.142
Uncertainty:	$\pm 0.041$	$\pm 0.050$	$\pm 0.104$
Ref. Std. Dev:	$\pm 4.5\%$	$\pm 3.3\%$	$\pm 4.9\%$
No. of samples:	1424	783	1086



PAS-500 Micro Air Sampler

# Atmospheric PFT Background Measurements at Four Nuclear Stations (March 2003)<sup>a</sup>

Station	Room	Area Ratio to ptPDCH		
		pc/mt	PECH	mc
North Anna	SWGR (U2 RA)	2.500	0.766	2.199
North Anna	MCR (U1 RA)	2.439	0.768	2.151
Surrey	SWGR (U2 RA)	2.474	0.819	2.186
Surrey	MCR (U2 RA)	2.447	0.805	2.153
Point Beach	Comp. Room	2.485	0.779	2.207
Point Beach	MCR	2.419	0.758	2.131
Kewaunee	MCR	2.488	0.784	2.190
Kewaunee	Equip. Room	2.496	0.799	2.237
		Average	2.468	0.785
		Rel. Std. Dev.	± 1.2%	± 2.7%
		n = 8		± 1.6%
Salt Lake City <sup>b</sup>	Average	2.442	-	2.142
	Rel. Std. Dev.	± 3.9%	-	± 4.9%
	n	~800	-	1086

<sup>a</sup> Control room samples were between 15 to 25 L of air.

<sup>b</sup> Salt Lake City samples were about 0.5 L of air.

# Fractional Penetration\* of Ambient Air PFTs Through NCS Corp. Charcoal

-- Testing at 23°C and 1.91 cfm through 2.07-in diameter test cell –  
(equivalent to 24.7 m/min or 120% of maximum typical plant use)

Time from Flow start	Duration Test	Test Port	0.49 atm** PDCB	0.047 atm					
				0.44 PMCP	0.142 PMCH	pc/mt	PECH	mc	pt
2.2 h	1.8 h	Meas. 1– in.	6.5	0.362	0.0646	0.0178	0.0147	0.0165	0.0115
		Calc. 2– in.	--	0.131	0.0042	0.0003	0.0002	0.0003	0.0001
		Meas. 2– in.	15.7	0.086	0.0130	0.0036	0.0065	0.0037	0.0078
5.3 h	4.3 h	Meas. 1– in.	1.9	0.385	0.0653	0.0159	0.0151	0.0153	0.0157
		Calc. 2– in.	--	0.148	0.0043	0.0003	0.0002	0.0002	0.0002
		Meas. 2– in.	3.7	0.071	0.0029	0.0006	0.0001	0.0004	0.0010
20 h	3.5 h	Meas. 1– in.	4.9	0.524	0.0736	0.0289	0.0290	0.0268	0.0271
		Calc. 2– in.	--	0.275	0.0054	0.0008	0.0008	0.0007	0.0007
		Meas. 2– in.	11.4	0.161	0.0056	0.0041	0.0019	0.0038	0.0004
48 h***	4.2 h	Meas. 1– in.	3.1	0.716	0.118	0.0329	0.0299	0.0312	0.0304
		Calc. 2– in.	--	0.512	0.0138	0.0011	0.0009	0.0010	0.0009
		Meas. 2– in.	8.0	0.349	0.0067	0.0014	0.0019	0.0010	0.0016
72 h***	4.1 h	Meas. 1– in.	3.5	0.838	0.127	0.0339	0.0318	0.0322	0.0268
		Calc. 2– in.	--	0.702	0.0160	0.0011	0.0010	0.0010	0.0007
		Meas. 2– in.	9.3	0.641	0.0094	0.0040	0.0052	0.0007	0.0042

\* Dual 1-in test cells and procedures were according to ASTM D 3803-91 (Standard Test Method for Nuclear-Grade Activated Carbon).

\*\* Vapor pressure at 25°C of each PFT, atm

\*\*\* Over 48 to 72 h, calculated penetration for 4 low vapor pressure PFTs averaged 0.097 ± 0.051%.

# Fractional Penetration\* of Ambient Air PFTs Through NCS Corp. Charcoal

- Testing at 23°C and 9.6 m/min (~47% of typical cell face velocity)
- Charcoal use: 7.4 days in end December 2002 and 1.4 days for this end-May 2003 testing

Test	Duration	Test Port	B.P. ~46° C**		76° C	102° C			
			PDCB	PMCP		pc/mt	PECH	mc	pt
1	7.0 h	Meas. 1-in.	3.0	0.56	0.091	0.0224	0.0220	0.0212	0.0188
		Calc. 2-in.	--	0.31	0.008	0.0005	0.0005	0.0005	0.0004
		Meas. 2-in.	4.2	0.26	0.004	0.0011	0.0010	0.0012	0.0014
2	8.2 h	Meas. 1-in.	2.3	0.52	0.092	0.0241	0.0239	0.0234	0.0213
		Calc. 2-in.	--	0.27	0.009	0.0006	0.0006	0.0006	0.0004
		Meas. 2-in.	3.4	0.26	0.004	0.0008	0.0015	0.0008	0.0009
3	7.0 h	Meas. 1-in.	2.0	0.51	0.106	0.0271	0.0249	0.0257	0.0232
		Calc. 2-in.	--	0.33	0.011	0.0007	0.0006	0.0007	0.0005
		Meas. 2-in.	2.9	0.29	0.005	0.0008	0.0009	0.0009	0.0012
4	4.7 h	Meas. 1-in.	1.8	0.52	0.103	0.0270	0.0281	0.0248	0.0227
		Calc. 2-in.	--	0.27	0.011	0.0007	0.0008	0.0006	0.0005
		Meas. 1-in.	2.6	0.27	0.004	0.0004	0.0006	0.0002	0.0005

\* Dual 1-in test cells and procedures were according to ASTM D 3803-91 (Standard Test Method for Nuclear-Grade Activated Carbon).

\*\* Boiling points of 300-, 350-, and 400-molecular weight PFTs, respectively

\*\*\* Over all 4 tests, calculated penetration of 4 high-boiler PFTs averaged **0.057 ± 0.012%**

# ATD Determination of Intentional Unfiltered In-Leakage in Charcoal Cell Test Apparatus

Tested at 23° C and 9.6 m/min (~47% of typical cell face velocity)

Test	$R_{fsa}$ , L/min	Rate of Unfiltered In-Leakage ( $R_{ui}$ ), L/min						Intentional Leak, L/min
		pc	PECH	mc	pt	Avg ± SD	net	
1	22.6	0.69	0.22	0.83	0.27	0.50 ± 0.30	---	None
2	22.3	1.19	1.06	1.84	1.25	1.33 ± 0.35	0.83 ± 0.46	0.75 ± 0.02
3	19.7	4.17	4.07	4.84	4.34	4.35 ± 0.34	3.85 ± 0.45	3.72 ± 0.03
4	21.7	1.79	1.70	2.18	1.82	1.87 ± 0.21	1.37 ± 0.37	1.45 ± 0.02

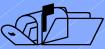
Proportioned to Full-Scale Rates, cfm

Test	Filtered SA	Unfiltered In-Leakage	
		Net Meas.	Set
1	2260 ± 23	--	None
2	2230 ± 22	83 ± 46	75 ± 2
3	1970 ± 20	385 ± 45	372 ± 3
4	2170 ± 22	137 ± 37	145 ± 2

# **Minimal Uncertainty Elements for ATD Measurements**

---

## Relevant uncertainty elements:

	<u>Typical Uncertainty</u>
 Volume of air sampled ... pumping rate times time	$\pm 3\%$
 Reproducibility of identical samples ... desorption and GC analysis	$\pm 5\%$
 Linearity of GC response ... from 50-L sample down to LOD	$\pm 2\%$
 GC limit of detection (LOD)	$\underline{\pm 1\% \pm 50 \text{ counts}}$ $\pm 6\% \pm 50 \text{ counts}$

# CR Unfiltered In-Leakage Rate by ATD

---

Rate ( $R_{UI}$ ) and Uncertainty ( $\Delta R_{UI}$ )

$$R_{UI} = R_{fSA} F_{dep} / (1 - F_{dep})$$

Where  $R_{fSA}$  = the total filtered supply air rate (cfm)

$F_{dep}$  = fraction of original PFT after depletion  
(PFT signal after/PFT signal before depletion)

Examples:

	1	2	3
$R_{fSA}$ (cfm)*	$1200 \pm 70$	$2000 \pm 140$	$4700 \pm 330$
$F_{dep}$	0.140	0.0150	0.0101
$R_{UI}$ (cfm)	$195 \pm 23$ (12%)	$30 \pm 4$ (14%)	$48 \pm 7$ (15%)

\* Separately determined by pitot traverses

# Control Room In-Leakage by ATD and AIMS Techniques

---

## Current Status

-  Audit by Dominion Energy of QAP
  - Week of 4 August 2003
  - New plan but built on BNL QAP
  - To meet the intent of 10 CFR 50 (Appendix B) and 10 CFR21
- 2. Proposed testing at Dominion Energy
  - North Anna                    AIMS                    Sept/Oct
  - Surry                          AIMS/ATD               Nov/Dec
  - Millstone                      ?                       Jan/Feb
- 3. Testing at Pilgrim Nuclear (Nov. 14-16)
  - Two 8-h days of ATD with simultaneous SF<sub>6</sub>
  - Third 8-h day of ATD, AIMS (1-zone), and SF<sub>6</sub>
  - Potential benchmark of 3 techniques simultaneously

# Special Issues Regarding Contracting to a National Laboratory

---

## Requirement to Work for Others (WFO) than DOE:

DOE encourages their resources be made available; however:

- ⌚ The techniques and/or tools must be unique and not commercially available
  - Both ATD and AIMS are unique techniques to BNL
  - The Sampling and analysis capability are unique to BNL
- 2. As a result of direct contact with licensees:
  - We can attend pre-bid meetings/discussions
  - We can submit unsolicited proposals (direct contact requests)
    - ... Before an RFP is issued
    - ... Based on our unique capability
- 3. BNL, as a GOCO research facility, cannot respond to RFPs
  - We cannot compete with private industry
  - If no responders are selected, we can submit an unsolicited proposal
- 4. BNL will eventually license to vendors or subcontract the capability

# **Unfiltered In-Leakage Quantified by PFT Measurements**

---

## Overview



Importance of new ATD method:

- a. provides direct precise determination of unfiltered in-leakages  
... with minimal uncertainty
- b. no tracer release required, no ceiling tiles removed, no mixing fans  
(as-is testing preferred)
- c. negligible intrusion in CR using only pocket-sized samplers
- d. applicable to pressurized and neutrally-balanced CRE with charcoal-filtered emergency air
- e. can provide indication of in-leakage locations directly into CR by using many samplers throughout CR
- f. sampling at charcoal-filtered SA grill will separately quantify unfiltered in-leakage in that system

# **Unfiltered In-Leakage Quantified by PFT Measurements**

---

## Overview



- Importance of new ATD method:** (continued)
  - g. Gives direct/average measure of estimated outside air-exposure concentrations at operator locations (eliminates concern for mixing, dead zones above ceiling tiles, etc.)
  - h. Sampling along the emergency ventilation system quantifies negative-pressure component in-leakage pathways (components outside the CRE):
    - ... Filtration/fan system housing and downstream duct work locations
    - ... CR AHU system housing and its upstream duct work locations
  - i. With 4 PFTs effectively depleted, gives replicate determinations for every sample
  - j. Intentional or incidental prior exposure of the charcoal to a PFT will preclude the method for that PFT
    - ... Fresh, laboratory-tested charcoal would restore the method for that PFT

# **Unfiltered In-Leakage Quantified by PFT Measurements**

---

## Overview

- ⌚ The importance of BNL-AIMS method:
  - a. quantify in-leakages from other tagged zones to facilitate mitigating strategies
  - b. no mixing fans (actually not desired)
  - c. negligible intrusion from miniature PFT sources and passive samplers
    - ... some testing will require using programmable BATS samplers
  - d. tagging and sampling strongly dependent on emergency system operations
  - e. better estimates of exposure dose at operator locations from other zonal PFT concentrations